Flux-lattice melting in $LaO_{1-x}F_xFeAs$: first-principles prediction

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We report the theoretical study of the flux-lattice melting in the novel iron-based superconductor $LaO_{0.9}F_{0.1}FeAs$ and $LaO_{0.925}F_{0.075}FeAs$. Using the Hypernetted-Chain closure and an efficient algorithm, we calculate the two-dimensional one-component plasma pair distribution functions, static structure factors and direct correlation functions at various temperatures. The Hansen-Verlet freezing criterion is shown to be valid for vortex-liquid freezing in type-II superconductors. Flux-lattice meting lines for $LaO_{0.9}F_{0.1}FeAs$ and $LaO_{0.925}F_{0.075}FeAs$ are predicted through the combination of the density functional theory and the mean-field substrate approach.

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I. INTRODUCTION

It is well known that the practical application of superconductors crucially depends on the high electricity transport without dissipation, so the searching for new superconductors with a high transition temperature has attracted considerable attention in material science. Recently, the novel superconductivity in iron-based layered superconductors has been reported, providing a new path to the high temperature superconductivity. The $LaOFeAs$ under doping with F^- ions at the O^{2-} sites was first discovered to exhibit superconductivity with $T_c = 26K$ by Kamihara et al [1]. Then the holes were introduced in $LaOFeAs$ by partially substituting La with Sr, the superconductivity was observed with $T_c = 25$ K [2]. Shortly after these studies, T_c was pushed up surprisingly to above 40 K when La in $LaO_{1-x}F_xFeAs$ was substituted by other rare earth elements [3, 4, 5]. In later experiments, the maximum T_c at about 55 K was achieved in $SmO_{1-x}F_xFeAs$ [6, 7] and $Gd_{1-x}Th_xOFeAs$ [8]. Though these superconductors have a high critical temperature, the mechanism and the possible difference compared with copper-based high-T_c superconductors are still very controversial. Obviously, more experimental and theoretical work are needed to understand the type-II superconductivity as well as the microscopic mechanism in these new iron-based superconductors.

Since the vortex-lattice solid (glass) state without linear resistivity is crucial for the application of high-T_c superconductors, the melting of the flux-lattice in bulk type-II superconductors is of fundamental and practical significance [9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19]. The elastic theory combined with the phenomenological Lindemann criterion has been used to study the flux-lattice melting in layered superconductors[20], which is based on the instability of the crystal. However, the regular flux-lattice structure loses simultaneously in the melting, the information for vortex liquid is then lacking, so the Lindemann criterion has drawback in the study of the vortex melting. The density functional theory has been successfully used in the flux-lattice melting of layered superconductors[9, 14, 17, 18, 19]. Due to the large anisotropy in these superconductors, weak Josephson interaction between layers can be neglected, both the interplane and intraplane interaction between the pancake vortices are electromagnetic in origin. The density functional method is usually well appropriate for layered superconductors with large anisotropy. Recently, a rather large anisotropy ~ 15 [21] was found in $LaO_{0.9}F_{0.1}FeAs$ and $LaO_{0.925}F_{0.075}FeAs$ (1111-type) by a bond structure calculation. The strong anisotropy in these systems was also confirmed recently in experiments[22].

In this work, we study the flux-lattice melting in these two samples in the framework of the density functional theory and the mean-field substrate model proposed recently [23]. In our first-principles calculations, the input contains just a few parameters. The two-dimensional (2D) one-component plasma (OCP) pair distribution functions (PDF), 2D OCP static structure factors (SSF) and 2D OCP direct correlation functions (DCF) are calculated in the celebrated Hypernetted-Chain (HNC) closure. The validity of the Hansen-Verlet freezing criterion is analyzed. The melting lines for $LaO_{0.9}F_{0.1}FeAs$ and $LaO_{0.925}F_{0.075}FeAs$ are plotted by using the mean-field substrate approach.

The paper is organized as follows. Section II describes the calculation of 2D OCP PDF. In section III, we analyze the Hansen-Verlet freezing criterion and present the calculation of flux-lattice melting lines. A short summary is given in the last section.

II. CALCULATION OF PAIR DISTRIBUTION FUNCTIONS

The electromagnetic vortex-vortex interaction is given by a three-dimensional (3D) anisotropic pair potential.

In Fourier space, it reads [9, 18]

$$\beta V(\mathbf{k}) = \frac{\Gamma\lambda_{ab}^2(T)(k_\perp^2 + (4/d^2)\sin^2(k_z d/2)}{k_\perp^2[1 + k_\perp^2\lambda_{ab}^2(T) + (4\lambda_{ab}^2(T)/d^2)\sin^2(k_z d/2]}, \quad (1)$$

where $\Gamma = \beta d\phi_0^2/4\pi\lambda_{ab}^2(T)$ and $\beta = 1/k_B T$, $k_\perp$ and k_z denote the component of $\mathbf{k}$ perpendicular and parallel to the c-axis respectively, $\lambda_{ab}(T) = \lambda_{ab}(T=0)/[1 - (T/T_c)^4]^{1/2}$ is the planar penetration depth, $\phi_0 = hc/2e$ is the flux quantum and d is the layer spacing between FeAs layers. In our calculation, d is twice lattice parameter c, i. e. $d \approx 17.5$ Å[1, 21]. According to Eq. (1), the intralayer potential is repulsive while the interlayer potential is attractive. Some parameters measured in a muon spin relaxation study [24] are used in the present work , e. g. $\lambda_{ab}(T=0) = 2540$ Å, $T_c = 26$ K for $LaO_{0.9}F_{0.1}FeAs$ and $\lambda_{ab}(T=0) = 3640$ Å, $T_c = 22$ K for $LaO_{0.925}F_{0.075}FeAs$. For convenience, the mean inter-particle spacing is taken as the length unit in the following.

In the classical liquid theory, PDF is defined as [25]

$$g(r, r') = \frac{\rho(r, r')}{\rho(r)\rho(r')}, \quad (2)$$

where $\rho(r)$ represents the probability to find a particle at r, $\rho(r, r')$ is the probability to find two particles at r and r' respectively, and the pair correlation function $h(r)$ is defined by $h(r) = g(r) - 1$.

In the uniform and isotropic liquid, the Ornstein-Zernike relation can be written as [25]

$$h(\mathbf{r}) = C(\mathbf{r}) + \rho\int d\mathbf{r}' C(|\mathbf{r} - \mathbf{r}'|)h(\mathbf{r}'), \quad (3)$$

while in the Fourier space, it becomes

$$h(\mathbf{k}) = C(\mathbf{k}) + \rho C(\mathbf{k})h(\mathbf{k}). \quad (4)$$

The HNC closure is widely used in the vortex system of type-II superconductors [9, 14, 18]. In this closure, the bridge function is set to zero and the DCF can be written as

$$C(r) = exp[-\beta V(r) + Y(r)] - Y(r) - 1, \quad (5)$$

where $Y(r) = g(r) - C(r) - 1$. Following the method proposed in Ref. [9], we extract the correlation functions by splitting $C(r)$ and $Y(r)$ into a short- and a long-ranged part as

$$C^n(\rho) = C_s^n(\rho) + C_l^n(\rho) \quad (6)$$

$$Y^n(\rho) = Y_s^n(\rho) + Y_l^n(\rho), \quad (7)$$

here $Y_l^n(\rho) = \beta V(\rho, nd)$, $C_l^n(\rho) = -\beta V(\rho, nd)$ and $C_s^n(\rho) = C_s^0(\rho)\delta_{n,0}$, ρ denotes the in-plane coordinate and n is the layer index [9]. In this way, the HNC equation in the short range part can be written as

$$C_s^0(\rho) = exp[Y_s^0(\rho)] - Y_s^0(\rho) - 1. \quad (8)$$

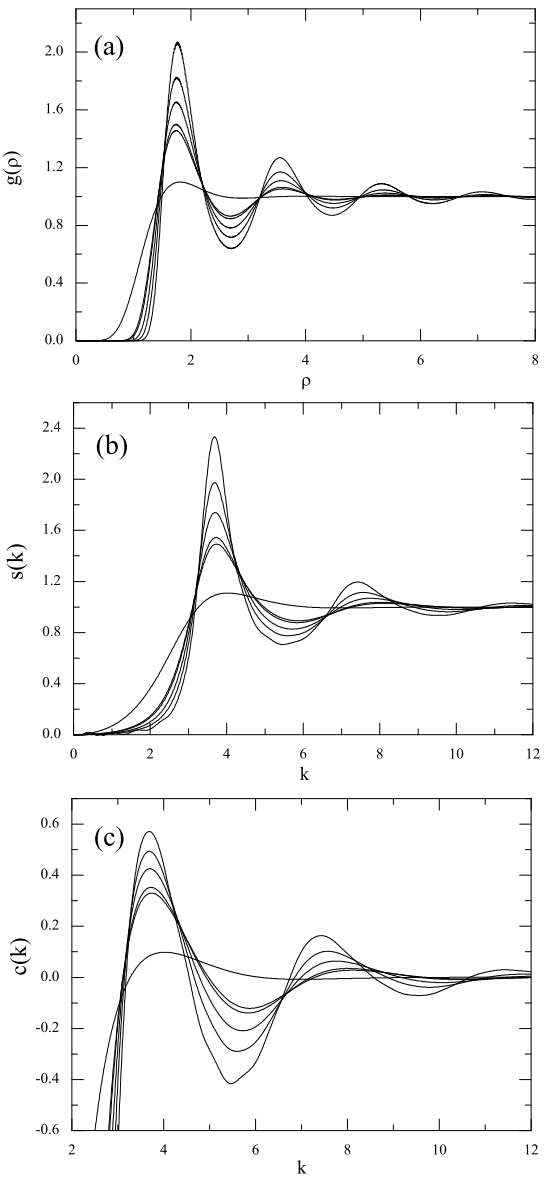

FIG. 1: (a) 2D OCP PDF (b) 2D OCP SSF and (c) 2D OCP DCF for $LaO_{1-x}F_xFeAs$ (x=0.1,0.075) at different temperatures. In all figures, from top to bottom at the first peaks: 10K for $LaO_{0.9}F_{0.1}FeAs$, 14K for $LaO_{0.9}F_{0.1}FeAs$, 10K for $LaO_{0.925}F_{0.075}FeAs$, 20K for $LaO_{0.9}F_{0.1}FeAs$, 14K for $LaO_{0.925}F_{0.075}FeAs$, and 20K for $LaO_{0.925}F_{0.075}FeAs$.

The in-plane PDF is then given by

$$\begin{aligned} g^0(\rho) &= C^0(\rho) + Y^0(\rho) - 1 \\ &= C_s^0(\rho) + Y_s^0(\rho) - 1 \\ &= exp(Y_s^0(\rho)). \end{aligned} \quad (9)$$

Using an efficient algorithm proposed previously [26], we are able to calculate the short range part of 2D DCF and $Y(\rho)$ successfully and then obtain the in-plane PDF. These functions are tabulated in a discrete form at about 1200 points. Since the PDF decreases rapidly in the or-

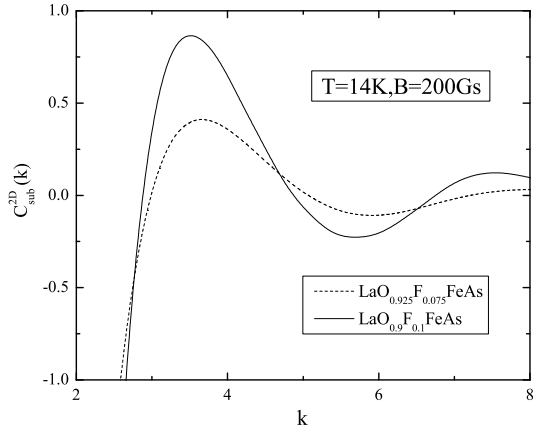

FIG. 2: DCF based on the mean-field substrate model for $LaO_{1-x}F_xFeAs$ (x=0.1,0.075) at T=14K, B=200Gs.

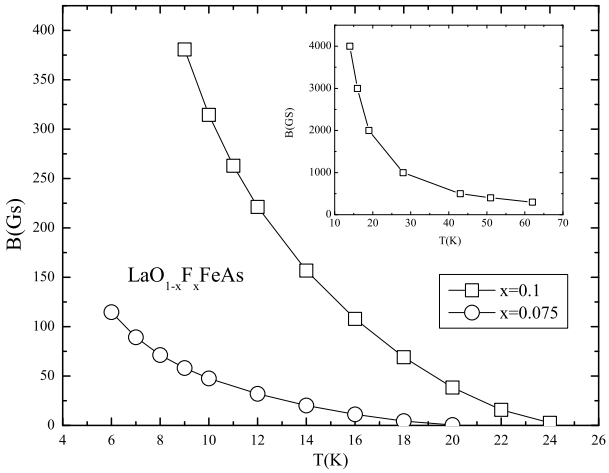

FIG. 3: Flux-lattice melting lines for $LaO_{1-x}F_xFeAs$. Inset: Flux-lattice melting line for $BSCCO$ calculated in Ref. [18].

der of the mean inter-particle spacing, they can be cut off in the real space to simplify the calculation. The criterion for this cutoff is that the converged functions are not influenced. In our calculation, all of the converged functions are stable and the relative difference is less than 10^{-9}. Actually, the value of PDF saturates at high field, consistent with the well-known fact that the vortex system decouples to a 2D system at high field. These PDF can be regarded as the 2D OCP one, which is shown in Fig. 1(a) for $LaO_{1-x}F_xFeAs$ (x=0.1, 0.075). It displays an oscillating behavior and becomes very small at large ρ.

III. FREEZING CRITERION AND MELTING LINES

In the classical liquid theory, the SSF is defined as

$$S(\mathbf{k}) = 1 + \rho \int d\mathbf{r}(g(r) - 1)exp(i\mathbf{k} \cdot \mathbf{r}). \qquad (10)$$

To provide a visual representation of the vortex structure, we calculate the 2D OCP SSF according to Eq. (10). The 2D OCP DCF are then easily achieved with the relationship $S(k) = 1/(1-C(k))$. Both 2D OCP SSF and 2D OCP DCF are shown in Figs. 1(b) and (c). In fact, the maximum values of these functions are found to decrease as temperature increases, shedding an insight of the 2D flux-lattice system in the melting process. Furthermore, all these functions for $LaO_{0.9}F_{0.1}FeAs$ exhibit a larger oscillation than those for $LaO_{0.925}F_{0.075}FeAs$ at the same temperatures, indicating that the 2D lattice structure in $LaO_{0.9}F_{0.1}FeAs$ is more clear than that in $LaO_{0.925}F_{0.075}FeAs$.

In the density functional approach first suggested by Ramakrishnan and Yussouff [27], the free energy function is given by

$$\begin{aligned} \beta\triangle\Omega &= \int d\mathbf{r}[\rho(\mathbf{r})ln\frac{p(\mathbf{r})}{\rho_l} - \delta\rho(\mathbf{r})] \\ &- \frac{1}{2}\int dr \int dr^{'} C(|\mathbf{r} - \mathbf{r}^{'}|) \\ &\times [\rho(\mathbf{r}) - \overline{\rho}^{3D}][\rho(\mathbf{r}^{'}) - \overline{\rho}^{3D}], \end{aligned} \qquad (11)$$

here $\rho(\mathbf{r})$ is the density at $\mathbf{r}$, $\overline{\rho}^{3D}$ is the three-dimensional homogenous liquid density, and $C(|\mathbf{r} - \mathbf{r}^{'}|)$ is the two-body fluid phase DCF. The two-body interactions between the particles are expressed in the second integral on the right hand side.

Recently, a mean-field substrate model was proposed to investigate the thermodynamic properties in layered superconductors [23]. Here we describe it briefly for completeness. Since the interlayer interaction between vortices is much weaker than the intralayer one in the strongly anisotropic superconductors, the vortices can be regarded as a collection of pancake vortices. In this way, the interplane and out-of-plane interaction could be separated and the DCF can be written as [17]

$$C_z(\rho) = dC^{2D}(\rho)\delta(z) - \frac{V_z(\rho)}{T}, \qquad (12)$$

where $V_z(\rho)$ is the out-of-plane interaction potential. In Fourier space, the effective DCF of the mean-field substrate model takes the following form

$$C^{2D}_{sub}(K) = C^{2D}(K) - \int dz \frac{\overline{\rho}V_z(K)}{Td}. \qquad (13)$$

We employ the above equation to extract the DCF based on the substrate model at different temperatures and magnetic fields. For instance, we present the DCF at T=14K and B=200Gs in Fig. 2. As proposed in Ref. [17], the single order parameter $C^{2D}_{sub}(G) = 0.856$

$((G = 8\pi^2\overline{\rho}/\sqrt{3})^{1/2})$ can be used to determine the flux-lattice melting lines, which is equivalent to the nontrivial solution for $\beta\triangle\Omega = 0$. On the other hand, the Hansen-Verlet freezing criterion is successfully used in the classical liquid theory [28]. This criterion states that the liquid freezes when the first peak of the structure factor reaches a critical value. The density functional theory in the vortex system of type-II superconductors is also based on the classical liquid theory, thus it is necessary to analyze the validity of the Hansen-Verlet freezing criterion in this issue. Actually, the single order parameter $C_{sub}^{2D}(G) = 0.856$ is just relative to the 2D Hansen-Verlet freezing criterion through the equation $S(k) = 1/(1-C(k))$. For the vortex system in layered superconductors with a large anisotropy, the liquid freezes when the structure factor at $k = G$ reaches the value 6.94. Interestingly, such a value is well consistent with the value of S(k) at low-field regime along the melting line obtained in previous work [18].

Finally, we employ the freezing criterion described above to locate the melting lines. Critical fields for $LaO_{1-x}F_xFeAs$ $(x = 0.1, 0.075)$ at different temperatures are calculated, the melting lines are shown in Fig. (3). For comparison, the melting line for $BSCCO$ obtained in Ref. [18] is also given in the inset. One can observe that the monotonic meting lines for $LaOFeAs$ are qualitatively alike but much lower than that for $BSCCO$. Recently, a quite large vortex liquid area in $LaO_{0.9}F_{0.1}FeAs$ compounds is observed in an experimental study based on the measurement of Nernst signal [29], consistent with the melting lines predicted in the present work. Interestingly, we find that in $LaOFeAs$ samples the melting line is changed substantially with the variation of F^- content, the melting line for $LaO_{0.9}F_{0.1}FeAs$ is much higher than that for $LaO_{0.925}F_{0.075}FeAs$.

The very recently discovered ternary iron-arsenides $(Ba, K)Fe_2S_2$ (122-type)[30] were suggested to be nearly isotropic[31]. The present method to locate the melting lines of course can not be applicable to these superconductors with low anisotropy.

IV. SUMMARY

The methodological difference between the present work and the previous study in Ref. [17] lies in the determination of 2D OCP direct correlation functions. In Ref. [17] these functions were obtained from Monte-Carlo simulations, while they are achieved *ab inito* from the microscopic vortex interaction in the present study. Therefore, the present approach is more intuitive, and the results are obtained without statistical errors like in Monte-Carlo simulations. The method adopted in the present work is also different from that in Ref. [9] where the density functional theory was first extended to the flux-lattice melting. A consistency to the Clausius-Clapeyron relation [17] can be reached with the use of the mean-field substrate model in the present work, but can not be obtained in Ref. [9].

We use the density functional theory and the mean-field substrate model to investigate the flux-lattice melting in iron-based superconductors $LaO_{1-x}F_xFeAs(x = 0.1, 0.075)$. The 2D OCP PDF, 2D OCP SSF and 2D OCP DCF for $LaO_{0.9}F_{0.1}FeAs$ and $LaO_{0.925}F_{0.075}FeAs$ are calculated at different temperatures via the HNC closure and an efficient algorithm. It is shown that the melting lines are quite low, well consistent with an experimental study on the Nernst signals [29]. Furthermore, the melting line is observed to change considerably with the modulation of F^- content in F-doped $LaOFeAs$ compounds, indicating that the interaction between vortices crucially depends on the F^- content. More experimental studies are needed to test the present theoretical predications.

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